AD NUMBER

AD813342

NEW LIMITATION CHANGE

TO

Approved for public release, distribution unlimited

FROM

Distribution authorized to U.S. Gov't. agencies and their contractors; Administrative/Operational Use; APR 1967. Other requests shall be referred to the Army Physical Research Laboratory, Attn: SMUEA-TSTI-T, Edgewood Arsenal, MD 21010.

AUTHORITY

USAEA, per USAEA notice, dtd 26 Aug 1971

AD 813342

AD

GCA TECHNICAL REPORT NUMBER 67-2-G

AEROSOL DISSEMINATION ASSESSMENT

First Annual Summary Report

by

R. Dennis
A.W. Doyle
D. Gordon

R.A. Gussman
C.O. Hommel
A.M. Sacco

April 1967

PHYSICAL RESEARCH LABORATORIES
RESEARCH LABORATORIES
Edgewood Arsenal, Maryland 21010

Contract DA-18-035-AMC-376(A)

GCA CORPORATION
GCA TECHNOLOGY DIVISION
Bedford, Massachusetts

GCA TECHNICAL REPORT NUMBER 67-2-G

AEROSOL DISSEMINATION ASSESSMENT

First Annual Summary Report

by

R. Dennis
A.W. Doyle
D. Gordon
R.A. Gussman
C.O. Hommel
A.M. Sacco

April 1967

This document is subject to special export controls and each transmittal to foreign nationals or foreign governments may be made only with prior approval of the CO, Edgewood Arsenal, ATTN: SMUEA-TSTI-T, Edgewood Arsenal, Maryland 21010

Physical Research Laboratories
RESEARCH LABORATORIES
Edgewood Arsenal, Maryland 21010

Contract DA-18-035-AMC-376(A)
Project 1B522301A081

GCA CORPORATION
GCA TECHNOLOGY DIVISION
Bedford, Massachusetts

FOREWORD

The work described in this report was authorized under Project 1B5223o1A081, Chemical Agent Dissemination (U). The work was started in June 1965 and completed in June 1966 for this reporting period.

Notices

Reproduction of this document in whole or in part is prohibited except with permission of the CO, Edgewood Arsenal ATTN: SMUEA RPR Edgewood Arsenal, Maryland 21010; however, DDC is authorized to reproduce for United States Government purposes.

The information in this report has not been cleared for release to the general public.

Disclaimer

The findings in this report are not to be construed as an official Department of the Army position, unless so designated by other authorized documents.

Disposition

When this document has served its purpose, DESTROY it.

DIGEST

The basic program objectives are to develop, to design, and to fabricate a complete aerosol assessment system. These were reviewed in terms of the capability of the instrumentation for the measurement of the mass concentration of active agent, concentration of degraded agent and inert materials, descriptive particle-size parameters for active agents, shape factor for solid particles, and bioeffectiveness. Sampling techniques were reviewed; e.g., filtration, inertial collection, scrubbing and absorbing, electrostatic, thermal percipitator, sedimentation, interpreted in terms of their applicability to the system. Special attention was given to proposed instruments for particle-size assessment. Several analytical methods for mass assessment are discussed including wet-chemical techniques and instrumental methods.

Results of experiments in a 1 m^3 test chamber are presented and discussed in such terms as floor sampling, an automatic particle counter, a beta absorption mass meter, a porous electrode collector, particle-decay analysis, a viewing microscope, mean decay of saccharin aerosols, sample size, location and time, and representative sampling.

TABLE OF CONTENTS

AEROSOL DISSEMINATION ASSESSMENT

I. INTRODUCTION

Basic program objectives outlined in previous quarterly reports (Ref. 1, 2, 3) are restated here for immediate reference.

The objectives of this program are to develop, design, and fabricate a complete chamber aerosol assessment system. The function of this system is to provide, by the selection and integration of appropriate instruments and analytical methods, a detailed description of chamber aerosols from as near as possible to the instant of dissemination to time periods up to one hour. By continuous or incremental sampling, the state of the aerosol should be definable over time intervals as small as one second.

Since the system is to be used as a major tool in the development of chemical devices, it should be adaptable to the assessment of a broad range of disseminators and agents and to most test chambers now used by groups within Edgewood Arsenal and by contractor organizations. Furthermore, extraneous effects attributable to chamber geometry, aerosol mixing, wall losses, electrostatic charge, and agglomeration should not interfere with proper assessment of disseminator functions.

For a broad capability in aerosol assessment, component instrumentation should permit the following measurements: mass concentration of active agent, concentration of degraded agent and inert materials, descriptive particle-size parameters for active agents, and shape factor for solid particles. In addition to a broad range of particle size, from less than 1 to 100 microns, particle dimensions should be relatable to those describing biological effectiveness of aerosol clouds.

In addition to the requirement that system components should define aerosol cloud properties described above, it is desirable that sensor outputs from these instruments should be amenable to direct display and/or data processing and storage. The extent to which data are displayed or processed and stored for later recall will depend upon which instruments or combinations thereof best meet the system objectives. In certain instances, automation may be highly impractical on the basis of the cost of the process and the accuracy with which measurements can be made with available equipment. Since the aerosol assessment system is to be furnished as a tested, prototype assembly at the end of a three-year period, the contract framework precludes extensive exploration of new or novel instrumental concepts. Therefore, it has been recommended that commercially available equipment or well-advanced experimental devices be used as the basic system components and that these components be modified and calibrated in accordance with specific system needs.

This approach implies that the extent to which ultimate program objectives may be realized is closely related to the present state-of-the-art of aerosol assessment. Thus, the thoroughness of the information survey, i.e. general literature search, review of DOD and contractual research programs, and contact with instrument manufacturers, is of particular importance to this program.

II. PROGRAM REVIEW

All phases of the research described in Quarterly Reports (Ref. 1, 2, 3) are summarized in this First Annual Report. Activities during the last quarter are reviewed in more detail in Section IV, particularly where they relate to a completed part of our studies.

During the first six months of the program, our major effort was an information survey which entailed a review of pertinent classified and open literature, site visits to Edgewood Arsenal, attendance at technical meetings, and communication with manufacturers and users of instruments considered as candidate components for the aerosol assessment system. Concurrently, the designs were completed and the construction begun on test apparatus considered basic to the program, regardless of final instrument selection.

A list of completed test apparatus with reference to the report in which a full description is given follows:

(1) A 1 m^3 cylindrical test chamber (4 feet diameter) with side and bottom entry ports for several types of sampling probes, three variable-speed, variable-orientation mixing fans, and a venting and filtered air supply system (Ref. 3).

(2) A special diluter and flow metering unit to convey sampled chamber air to a light scattering particle counter located immediately below the bottom of the chamber. Provision is made to collect effluent air from the viewing cell of the particle counter on membrane filters to assist in calibration (Ref. 2 and 3).

(3) An in-line cascade impactor with easily removed sampling slides and an inlet diluter section to provide better control of particle density on collection slides (Ref. 3).

(4) Sampling probes and flow metering units for simultaneous sampling at three elevations within the test chamber. These include a miniaturized filtration unit for high velocity sampling and an iris-shuttered unit to evaluate aerosol deposition during nonsampling periods (Ref. 3).

(5) Collection media including all-glass, cellulose, and membrane filters as well as glass slides to collect the aerosols produced in the one cubic meter chamber. Tests were conducted to evaluate which media or combination of media interfered with the analysis techniques (UV spectroscopy, gravimetric weighing, and light microscopy).

(6) Aerosol generation equipment for metered dispersion of mono-and polydisperse solid and liquid particles. Included are compressed gas ejectors for dry powders, pneumatic atomizing nozzles, and evaporation-condensation methods (R... 3).

Activities involving preliminary calibration of test equipment were also carried out. These included calibration of particle sizing instruments, i.e. single-particle light scattering unit and a GCA modification of the Battelle type cascade impactor versus direct microscopic examination of filter samples (Ref. 3).

Determination was made of factors leading to optimum stirring conditions by simultaneous sampling at various locations (Ref. 2 and 3). More information on this subject is presented in Section III.

During the second quarter and continuing throughout the year, methods of processing raw size measurement data to provide rapid description of aerosol size properties were studied. Computer programs were developed to generate synthetic decay curves for particle clouds of known composition. These data were applied mainly in testing reconstruction methods proposed for determining initial particle size distribution based upon analyses of mass decay curves. Inversion techniques which might be applied to mass decay curves are discussed in Reference 3.

Several chemical methods for agents and test materials were evaluated experimentally in conjunction with other GCA programs (Ref. 3). In addition to these reported findings, details of the methods and results are discussed further in Section IV C of this report.

Results of the information survey were broken down into three major areas (Ref. 1): (1) chemical methods; (2) chamber effects on test aerosols; and (3) optical instrumentation for determination of particle size parameters.

The chemical methods were reviewed in the Second Quarterly Progress Report (Ref. 2) under "Review of Methods for Quantitative Measurement of Chamber Aerosol Concentration, Section V." Special problems relating to chamber effects on test aerosols were discussed under "Representative Sampling," (Section III-H, Ref. 2).

The commentary and evaluations on optical sizing methods were essentially complete as reported in the First Quarterly Report (Ref. 1). It appeared rather early in the program, for example, that laser holographic or television camera methods were not sufficiently advanced for system application. No new information has been received which would lead to any change in the status of the devices mentioned above. The only optical devices considered as possibilities were single particle light scattering counters (Ref. 1, 2, 3). Unless these devices can be designed with the added capability of discriminating between active agent and debris particles, we do not recommend them as basic system components.

At the end of six months, a restatement of program objectives and a proposed listing of instrument selections were presented, based upon the information survey and discussions with Edgewood Arsenal personnel (Ref. 2). In this report, those program objectives relating specifically to particle sizing methods, e.g. characteristic diameter, distribution parameters, shape factor, uni-and/or multimodal properties, are reviewed in Section III-e. A revised tabulation of instruments proposed for particle size assessment of chamber aerosols are given in Table I.

A similar summary of those techniques for assessment of mass concentrations of chamber aerosols still under consideration are listed in Table II and discussed in Section III E. In addition to those analytical methods cited in Table II, several others were investigated, including the following:

(1) Raman spectroscopy

(2) Laser techniques for Raman and infrared spectroscopy

(3) Microwave spectroscopy

(4) Gamma ray attenuation

(5) Arc emission spectroscopy

(6) X-ray emission spectroscopy

(7) Attenuated total reflectance (ATR) spectroscopy

The reasons for rejection of these methods are discussed fully in Reference 2. Generally, these techniques either failed to meet sensitivity, specificity, or reproducibility requirements or were too costly for inclusion in the assessment system. It is suggested, however, that should significant breakthroughs be achieved with any one of the methods above, its use should be reconsidered.

Recent developments in both increased power and available wavelength of lasers suggest that these sources may indeed be of increased utility in both Raman and infrared spectroscopy. It is, however, clear that, at present, considerable development work would be required to realize the potential advantages of such systems.

Several sample collection methods listed in Table I were examined during the information survey. A detailed evaluation of potentially useful sampling methods was furnished in Reference 2.

Despite the apparently broad choice of instrumentation, very few of these devices are readily adaptable for chamber assessment applications. Aside from the fact that certain methods do not respond to a

TABLE I

REVIEW OF SAMPLING TECHNIQUES FOR CHAMBER APPLICATIONS

A. Filtration

1. Manually operated devices
2. Automatic devices

B. Inertial Collectors

1. Centrifugal types
2. Impingers
3. Cascade (Jet) impactors
4. Special impaction techniques

(a) Sensing wire grids
(b) Foil puncture

C. Scrubbing and Absorbing Techniques

D. Electrostatic Methods

1. AC and DC precipitation
2. Ion mobility

E. Thermal Precipitation

F. Sedimentation and Wall Deposition

1. Floor sampling trays
2. Conveyor belts

(a) Tray retrieval
(b) Continuous sampling and collection

3. Microbalance technique

G. Other Methods

1. Diffusion battery
2. Freezeout traps
3. Absorption properties
4. Differential conductivity

TABLE II

SUMMARY OF PROPOSED TECHNIQUES FOR ASSESSMENT OF MASS CONCENTRATIONS OF CHAMBER AEROSOLS

INSTRUMENTAL METHOD	DETECTION PRINCIPLE	READOUT TIME	AUTOMATION POTENTIAL	SPECIFICITY FOR AGENT	COMMERCIAL EQUIPMENT AVAILABLE	REMARKS
Autoanalyzer	Wetchemical processing. Conversion to colored compounds usually detectable by spectrophotometric analysis. Other sensors may be used.	Variable, few minutes to two hours. Depends upon reaction rates or specific treatments.	Excellent	Excellent	Yes	Adaptable to almost any colorimetric analysis. Reagents must not react with or corrode instrument components. Modular construction permits interchange of analytical equipment for analysis of different chemical agents.
Flame Photometer	Emission spectroscopy, detection of characteristic atomic lines of chemical agent compounds	Direct	Excellent	Poor specificity, excellent for undegraded agent. Otherwise no distinction between active and degraded material	Yes	Sample can be analyzed as vapor or as solvent solution. Adaptable to agents containing P, S, As, or other elements not present as interfering background.
Infrared Spectroscopy	Measures IR absorption based upon molecular properties.	Direct	Excellent	Excellent	Yes	Sample can be analyzed as vapor or as solvent solution. Use of more than one wavelength permits positive identification and minimizes effects of contaminants.

sufficiently large particle size spectrum (e.g., sensing wire grids, foil puncture, thermal precipitation, and diffusion battery) many of the devices are too fragile for effective operation in typical test chambers. Others would require major design modification which might entail a lengthy and not necessarily successful test program.

In terms of known performance and established dependability, dry filtration and wet impingement (scrubbing) are given priority. Furthermore, both techniques have been successfully automated for use in sampling systems for atmospheric pollutants in ambient atmospheres and industrial stacks. At this writing, it appears possible to design similar devices expressly for chamber applications, although the required research and development effort may be too extensive for the present contract. During the next quarter, it is expected that a positive decision will be made as to what constitutes an optimum degree of automation.

III. PROPOSED INSTRUMENT SELECTIONS FOR PARTICLE SIZING OF CHAMBER AEROSOLS

A. INSTRUMENT SELECTION AND DESCRIPTION

A restatement of program objectives was presented (Ref. 2) based upon the review of information sources during the first six months of the contract. Although no drastic changes were suggested, it was pointed out that certain practical compromises were considered necessary for successful pursuit of these studies. First, the number of instruments selected for fully or partially automatic and manual operation would be limited. Second, the number of measurements and the types of sensors would be governed by existing chamber design and operating conditions. Third, the precision required to furnish a useful description of chamber aerosol properties would be based upon the cost and complexity of proposed system instrumentation. It was stated that the third item, rather than the present state of technology, would probably be the controlling factor in the ultimate selection of system components.

The following four types of instruments and/or sampling techniques for particle size assessment were recommended for laboratory evaluation and study during the second half of the first year's program: (1) mathematical or graphical analyses of mass decay curves, (2) cascade impactors, (3) light scattering particle counters, and (4) direct observation of membrane filter samples.

Several other techniques, including holographic and TV camera imaging, electrostatic precipitation and fractionation, wire impaction, and centrifugal techniques related to particle inertia or momentum, and differential diffusion and conductivity methods were rejected on the basis of one or more of the following: primitive state of development, excessive cost, or incompatibility with chamber test procedures.

The four selected methods were evaluated in the light of the revised program objectives (Ref. 2) and performance requirements. Based upon known performance characteristics and the results of laboratory tests, it appears that the instrument types and techniques suggested previously still remain as optimum choices. Unfortunately, no single instrument or method among the four cited above can perform all of the sizing functions listed under the five major program objectives: (1) particle size measurement from 0.1 to 100 microns, (2) estimate size distribution parameters and characteristic dimensions of aerosol cloud components, (3) indicate multimodal properties (if present) of chamber aerosols, (4) estimate shape factor and relate to aerodynamic and surface properties of particles, and (4) estimate size distribution properties of initial aerosol as close to dissemination time as possible.

Based upon available instrumental methods and the present techniques for maintaining stirred settling conditions within test aerosol chambers,

it appears that no useful size measurements can be performed upon chamber clouds until 1 to 3 minutes of stirring have ensued following aerosol dissemination. This means that the only way in which the initial cloud parameters can be defined is through extrapolation of data obtained at a later time. Since the reliability of extrapolated data decreases the further one departs from the measured observations, techniques that furnish the complete size spectrum with a single short period sample should give the best results. Thus, instrument choice is important in meeting the time considerations of objective (5).

1. Mass Decay Curve Analysis

Mathematical and/or graphical methods are proposed, which may be used to infer the initial (time 0) particle size distribution of chamber aerosols. Simple graphical approaches have been described (Ref. 4) which involve the visual matching of experimental decay curves with theoretical decay curves generated from known size distributions, as assumed in this work to be a logarithmic-normal distribution. The graphical method usually provides only a rough estimate of size properties, although on limited occasions the curve fit is excellent. The main difficulty is that the assumption of log-normal properties is basically a matter of convention, and not necessarily true for many chanber aerosols. Although the estimates of mass median diameters may be reasonable, e.g. $\pm$ 20 percent accuracy, the fraction of the particles in specific size ranges may be grossly distorted. If such data were used to predict meteorological transport of the parent cloud, serious errors in predicted dosage rates might result.

More sophisticated methods involving the inversion of decay curve parameters to obtain the initial distribution have been studied by many investigators (Ref. 5 through 10). Regretably, no complete successes have been reported, although with certain restrictions the process has yielded satisfactory results for special situations. Various approaches cited in the literature and also under study at GCA are discussed in Reference 3.

At present, application of Phillips Method (Ref. 11), in which corrections are made for deviations from the ideal matrix structure, appears to show the most promise although its strength in delineating multimodal distributions is questioned. Since an effective solution to the inversion problem has not yet been formulated, we believe it essential to continue studies in this area. The main advantage of inversion is that measurement techniques are now available from which the basic inversion method parameters necessary can be determined. The collection of aerosol samples upon dry filters or in liquids for mass analyses alone is a simpler procedure than determining particle size distributions directly by cascade impactor, light scattering particle counters, and direct microscopy. Furthermore, sensor outputs from automatic analyzing equipment can be taped and stored for computer processing with readily available commercial

equipment. It is necessary, however, to accept a limitation in the range of sizes that can be defined by inversion techniques. Depending upon the volume (and particularly the height) of aerosol chambers, the smallest resolvable particle diameters are of the order of 2 to 3 microns for chambers 15 feet high. The upper size limit that can be determined by this method is approximately 100 microns, although sampling errors are expected for diameters greater than 50 microns. Small experimental chambers in the cubic meter catagory may permit size definition down to one micron. This points out that decay curve inversion techniques are insufficient to establish size parameters for sub-micron-sized aerosols of the fume or condensation types. From a practical viewpoint, this limitation is tolerable with most explosively generated solid or liquid aerosols, since at least 90 percent by weight of the parent cloud is composed of particles greater than 2 microns in diameter.

2. Cascade Impactors

The cascade impactor has been proposed as an intermediate sizing device for assessing particle sizes from 0.5 to 25 microns. We have described (Ref. 1, 2, 3) a prototype instrument designed by GCA in which many of the objectionable features found in commercial instruments are corrected. Although it is not feasible to develop a fully automatic cascade impactor under the present program, it is now believed that use of the impactor on a semi-automatic basis can provide valuable size assessment data. It was suggested that manual loading and unloading of collecting stages in combination with rapid automatic analysis could provide size distribution properties of the chamber aerosols at from 2 to 4 specific times during a test. Data from this source would complement data derived from inversion and, in the event of other instrument failures, would prevent loss of critical experiment data. Aside from recommending the impactor as a system component, its use as a calibration device is considered essential to the program. Preliminary tests of the GCA impactor design have shown good agreement between mass concentrations determined by parallel filter and cascade impactor samples. These data demonstrated that two major problems encountered with this instrument had been minimized, i.e., intrastage deposition of sampled material and representative sampling for both total mass and size properties. Based upon indicated performance, it is proposed that the impactor be considered as an alternate selection among the instruments or methods required to describe particle size distributions from 0.5 to 25 microns.

3. Light Scattering Particle Counter

The advantages and disadvantages of light scattering particle counters have been discussed at length in our previous reports. The equipment used in our laboratories operates or measures the amount of light scattered at a 90-degree angle (relative to the incident white light beam) by single particles. The intensity of the scattered light flux is approximately proportional to the particle projected surface area. With the use

of gating, logic, and discriminating circuitry, light pulses detected by a photomultiplier tube are sorted in accordance with predetermined size ranges (channels) and displayed on a counter. Provision is made for automatic or manual sequential counting and display. With a single channel read-out, approximately 0.32 min. is required to examine one size range. Hence, complete sizing and counting of an aerosol in which the particles fall within 15 size channels would require about 5 minutes. If a commercially available multichannel recording system were used, the time for complete size analysis would be reduced to 0.32 min. From an automation viewpoint, this type of instrument has much more to offer in that its sensor output can be readily transferred to magnetic tape and with modification it can be made to perform certain computer functions.

Our laboratory tests with a commercially available unit have shown that both particle number concentration and size-distribution data were in good agreement with simultaneous measurements made with the cascade impactor and by direct microscopic sizing. These results were obtained with a specially designed sampling and dilution section which was attached to the particle measuring chamber. Without this GCA modification, the light scattering counter was not suited for chamber applications. According to our laboratory tests, the working particle size range for the counter was from 0.5 to 50 microns, a considerably broader capability than that exhibited by the cascade impactor.

If chamber aerosols consisted of single component dispersions with respect to physical and chemical properties (e.g., generation by an atomizing nozzle at ambient temperature), properly designed light scattering particle counters would probably be the most useful tool for size assessment. It would only be necessary to calibrate the unit against aerosols of known properties to obtain the correct pulse amplitude size relationship. In practice, however, several test aerosols, particularly those generated by explosive means, are represented by heterogeneous systems in which large variations in particle shape, refractive index, absorbtivity, and surface properties can be expected. Current light scattering instruments do not have the capability of interpreting the above variations in any meaningful way with regard to the true-particle size characteristics. Therefore, it can be stated that existing types of light scattering particle counters can serve only in a limited capacity as a component in chamber aerosol assessment systems.

If an instrument were available which could distinguish between spherical and non-spherical particles, liquid droplets might be differentiated from irregularly shaped solid particles representing solid decomposition products, explosive debris, and casing fragments on the basis of symmetrical and asymmetrical light scattering paterns. Similarly, one might use two light exciting sources, one in the visible range and the other in the infrared, so that comparison of the ratios of signal intensities measured at a specified angle for absorbing and dielectric particles might permit differentiation between agent and non-agent materials.

Our present opinion is that a decision should be made soon, as to whether a research and development study should be implemented to determine the feasibility of designing a new light scattering device along the lines described above and in Reference 3. Since extensive research and development efforts were not considered as a part of this contract, justification would depend upon demonstrated inadequacies in other size assessment techniques currently available. It also appears logical to re-examine certain program objectives with regard to range of particle sizes of immediate interest and the acceptable precision or refinement desired in defining aerosol cloud properties.

4. Direct Observation of Membrane Filter Samples

Visual examination of particulate materials collected upon membrane type filters is proposed as a means of calibrating the various sizing devices to be used in the chamber assessment system. Although data obtained from this source affords the best description of particle size parameters, the lengthy analytical time required and the high degree of skill and patience demanded in examining the filters restrict this method to a calibration role.

Examination of solid particles is not expected to present any difficulties, since a variety of index of refraction matching liquids are available to make the membrane filter media transparent without dissolving the collected particles. Alternatively, a metallographic-direct reflecting optical system can be used without filter wetting.

Liquids which wet the surface either completely or partially cannot be collected on membrane filters since they tend to spread so that geometric description of size becomes impossible. For such materials, thermal or electrostatic precipitation might be used as alternate techniques. Unfortunately, no broad applications appear possible for the latter methods, since they have been developed mainly for the assessment of very fine (micron to submicron range) particles. These techniques were evaluative in Reference 2. On the basis of currently available sizing methods, the cascade impactor appears as the most practical instrument of choice for sizing liquid aerosols. Although measurements cannot be considered as refined as those obtained by microscope observation, they should provide adequate information to predict atmospheric transport properties of chemical agent dispersions. If required, one can also examine cascade impactor stages microscopically (a necessary calibration procedure) to improve data quality. Wetting problems encountered with some organic liquids can be overcome by using special plastic surfaces, e.g. Teflon or Tedlar.

B. PERFORMANCE REQUIREMENTS

1. Range of Particle Sizes of Interest

The information survey and a review of program objectives with Edgewood personnel indicated that the overall particle size assessment

capability of system instrumentation should bracket sizes from 0.5 to 100 microns. The dissemination techniques for this size criterion range from pyrotechnic generation of fume particles to hydraulic or pneumatic atomization of coarse droplets. The techniques discussed in this report were evaluated in chamber tests and provide size distribution data for the size ranges listed in Table III.

TABLE III

WORKING SIZE RANGE FOR CHAMBER AEROSOL ASSESSMENT METHODS

Sizing Method	Working Size Range Microns
Mass Decay Curve Analysis	2 to 100
Cascade Impactor	0.5 to 25
Light Scattering Particle Counter	0.5 to 50
Membrane Filter-Optical Sizing	0.5 to 100

2. Size Distribution Parameters and Shape Factors

Characteristic mean and median diameters for number, surface, and volume properties of polydisperse systems and their related distribution functions, i.e. arithmetic or geometric standard deviation, can be determined directly or indirectly by any of the sizing methods listed in Table III. The relationship of measured diameter to that which is related to the aerodynamic behavior of particles during meteorological transport or within the airways of the respiratory system was reviewed by the Assessment Working Group (Ref. 12).

Direct microscope examination of membrane filters or cascade impactor stages permits an estimate to be made of either a characteristic linear diameter, D_1, or a characteristic surface area diameter, D_2, depending upon the sizing technique. Averaging a linear dimension, in one direction only, is sufficient for the estimate of D_1 when particles are positioned randomly. If irregular particles are reduced visually to an equivalent cross sectional area (a standard technique used with the Porton graticule), the reported diameter, D_2, reflects total or projected area, provided that particle orientation upon the viewing surface is unbiased. Only in the case of spherical particles will D_1 and D_2 have identical values. Therefore, it is necessary to apply a correction (shape) factor to transpose from one measurement system to another.

If nonspherical solid particles are examined with a conventional microscope system, descriptive diameters D_1, a measure of length and D_2, a measure of projected or total surface area can be estimated directly. When the total mass of solids can be determined by gravimetric or chemical analysis and the particle number concentration and true particle density is known, a third diameter, D_3, often referred to as the diameter of the equivalent sphere, can be calculated.

For an irregular particle with the ratio of its longest to shortest diameter in the x, y, z directions not exceeding 1.5, preferential orientation on flat surfaces is not a significant factor. Hence, measurements with stage or ocular micrometers are unbiased.

Particle size classification by inertial separation, e.g. cascade impactor, provides a measure of size which relates to the aerodynamic behavior of a particle, which is a function of particle mass and is proportional to k D_3. If calibration of the cascade impactor is carried out with solid spherical particles, the estimated diameters, D_1 and D_2, which correspond to linear or area properties, are the same as those obtained by direct microscope observation. This is not the case when particle shape and density differ from those of the materials used in the instrument calibration. Therefore, correction factors must be used to compare observations with those made with other sizing devices.

Interpretation of apparent particle dimensions indicated by decay curves are subject to the same considerations applying to cascade impactor data. In both cases, measurements reflect particle size as a function of terminal settling velocity, V_t, which, in turn, is defined in terms of particle shape and density.

The particle dimensions of major concern, as stated previously, are those which relate to meteorological transport and biological assimilation. For this reason, particle size definition by inertial methods, e.g. mass decay curve or cascade impactor analysis, is ordinarily the most useful approach. Unless particle shape and density deviate extremely from those of solid or liquid spheres of the parent agent, the observation that a specified weight fraction of the cloud settles as if it were composed of 5 to 15 micron diameter solid spherical particles is sufficient to predict both aerodynamic transport and biological effectiveness.

If the particles are irregularly shaped, their mass will be larger than that for the spherical particles; thus a conservative estimation of dosage is made. At the same time, the maximum dimension of such a particle will exceed that of the spherical particle of equal mass. Unless this shape variation represents an extreme deviation from the spherical, however, the aerodynamic motion and ultimate deposition point of the particle within the respiratory tract will not be changed significantly. On the other hand, a loosely flocculated material may be large in overall dimensions relative to the size of the air paths of the lower respiratory tract despite the

fact that it possesses a low terminal settling velocity. If the biological action of the agent is highly specific insofar as tissue type is concerned, then it should be determined whether it is arrested by the nasal filtration system or makes a deeper penetration.

The sizing methods described here allow for the practical estimation of shape factors which can explain excursions in particle behavior when form and density effects cannot be neglected. At least two independent measuring procedures are required to determine effective shape factors: direct microscope observations of filter samples combined with total particule count and total particle mass; cascade impactor tests with combined optical sizing and mass analyses, and mass decay curves in combination with microscope observation of filter samples or cascade impactor stages.

Light scattering measurements might be considered as a substitute for microscope identification on the basis of particle projected area. It should be remembered, however, that failure of present light scattering instruments to distinguish between particles having different physical and/or chemical properties limits their use to single component aerosols.

3. Multimodal Distribution

Since there has been no change in our approach to the treatment of multimodal distributions from those described in Reference 2, the present discussion is limited to a few practical considerations.

First, the instruments proposed for use in the aerosol assessment system can provide a description of the aerosol cloud only during the periods of actual experimental measurements. Should bi- or multimodality in fact exist between the instant of cloud formation and the time of the first assessment measurement (perhaps 2 to 3 minutes), it could at best only be inferred and certainly not proven mathematically. Unless one has designed the aerosol generation system specifically to produce large particle coverage of very small areas, description of cloud size properties in the first two minute interval is of academic interest only. Generally, those particles which are in suspension after 2 to 3 minutes are of primary interest insofar as broad area coverage and inhalation are concerned.

Second, the sizing procedures proposed for system usage will show multimodal characteristics within reasonable bounds. Direct microscope observation of filters of impactor stages represents the best method for this purpose. It should be noted that apparent multimodal properties may be the result of statistical variations arising from limited data. Unless the function of cloud assessment is to provide information for mechanistic studies of dissemination processes, it appears that the presence or absence of deviations from unimodal distributions can only be judged by a combination of mass decay and cascade impactor analyses. The

argument advanced here is that multi-modal properties concealed between the characteristic size intervlas of a 5 to 6 stage impactor will have no important bearing on the behavior and the biological effectiveness of the cloud. Should a cumulative size distribution based upon impactor measurements show a strong inflection point more refined procedures would be recommended to establish the validity and the significance of the multi-modal properties.

4. Sample Size

The items discussed in the following paragraphs were reviewed at length in Reference 2. The results of experimental work and further literature review during the interim now allow more definitive operating conditions to be set for sampling systems, as well as a more definitive choice of sampling instrumentation to be made.

As indicated, the size of the sample would be controlled by: (1) The sample size in terms of number of particles or mass of material needed to furnish a measure of chamber concentration of predetermined statistical accuracy and (2) the minimum amount of agent which can be analyzed quantitatively by existing instrumental techniques.

It now appears that sampled air or gas volumes from 20 to 30 liters will provide sufficient agent for detection by the analytical methods described in this and earlier reports (Ref. 1,2,3). Therefore, we do not think it necessary to resort to the high volume (1,000 to 10,000 l/min) electrostatic precipitation as suggested by Lundgren and Graf (Ref. 13) and by Space General Corporation (Ref. 14). However, high volume filtration units should be used as standbys when unique agents or test materials can be analyzed most rapidly by simple gravimetric techniques. GCA has successfully used this procedure under another contract with Edgewood Arsenal [DA-18-108-AMC-249(A)]. Samples collected for one minute periods at flow rates of the order 1500 l/min provided enough material for gravimetric analysis.

5. Sampling Location

Several tests have been performed in our one cubic meter test chamber (Reference 3 and Section IV of this report) to establish optimum sampling locations. If it is assumed that uniform chamber mixing can be achieved, any location within the chamber should provide a representative sample. Results of fan stirring tests indicate that concentration uniformity should be obtainable. Furthermore, the stirred zone appears to extend very close to the chamber walls. Therefore, we believe that short sampling probe extensions within the chamber proper will be permissible. Under these conditions, the optimum operating conditions cited in Reference 2 can be realized, e.g. (a) minimum obstruction of chamber interior, (b) easier maintenance and shielding from explosive missiles, and (c) minimal length of sampling lines to analytical equipment located outside the chamber.

6. Sampling Time

(a) Relative to Dissemination Time

It has been shown that at least one and up to three minutes must be allowed between agent release and the time of initial sampling before meaningful data can be obtained. Stirring tests suggest that uniform mixing is attained sooner in small chambers than in large ones. The precise starting time should be based upon tests made in each field installation.

(b) Sampling Duration and Frequency

Tests indicate that samples collected over one-minute intervals will provide accurate representation of true cloud decay behavior in typical test chambers. The average cloud concentration over the sampling interval is essentially the same as the theoretical value corresponding to the averaged time of test.

Criteria for frequency of sampling remain as cited previously (Ref. 2) i.e., sampling at two-minute intervals throughout the test except for situations where decay is rapid (< 20 percent agent remaining in suspension at the end of five minutes). In the latter case, one-minute intervals between samples are proposed with sampling duration reduced to less than 1.0 minute.

7. Representative Sampling

Collection of representative samples of aerosolized particulate material from stirred chamber atmosphere has been discussed (Ref. 1,2,3). The results of simultaneous sampling at various locations within a one cubic meter test chamber showed that adequate mixing could be achieved insofar as mass concentration and size distribution were concerned. These tests, however, did not necessarily prove that sampling was non-selective as applied to particle diameters greater than 30 microns since identical sampling conditions prevailed at each station.

May (Ref. 15) and Levin (Ref 16) indicated that the problem of collecting all of the suspended particles originally suspended in the sampled gas volume has no simple solution. From a practical point of view, Levin suggests that high sampling velocities relative to average chamber circulation velocities minimize inertial fractionation (and loss) of large particles. Preliminary comparisons of simultaneous samples collected by dry filter and cascade impactor showed identical concentration. Since the sampling velocities at the surface of the filter and at the inlet to the cascade impactor were approximately 50 and 400 ft/min, respectively, one might conclude that the velocity factor is not important. These tests, however, were performed with saccharin aerosols

having a mass median diameter of about 18 microns. Possibly, the presence of a significant number of large particles, e.g., a 50 micron diameter mass median aerosol would have shown an appreciably higher concentration according to cascade impactor measurements. Equipment designed and described previously will allow for high velocity collection of filter samples (Ref. 2,3); therefore, the effect of sampling velocity with coarse particulate suspensions can now be studied.

IV. TESTING AND EXPERIMENTS DURING LAST QUARTER

A. CHEMICAL METHODS

Spectrophotometric analytical procedures for saccharin, di-2-ethyl hexyl phthlate, and agent CS were developed in the ultraviolet region. A Perkin-Elmer 4000A Spectracord was used. The samples were placed in 10 mm quartz cuvettes. The calibration curves are shown in Figure 1. It may be seen that they were linear over the concentration ranges.

After the samples were collected on filter paper, they were leached in 10 ml of methanol for 15 minutes. A 5 ml aliquot was withdrawn by a glass syringe, and filtered through a Millipore Syringe Filter, loaded with solvent washed Whatman No. 41 paper. The absorbance of each solution was determined immediately and the concentration calculated.

The filter paper used to collect aerosol samples in the one cubic meter test chamber was selected on the basis of least interference in the chemical determination. For example, methanol extracts of Millipore filter media showed a high absorbance in the 250-300 mu region, which lead to rejections of the filter. Washings from fired and solvent washed glass fiber paper (1106B) showed a variable background reading. Since solvent-washed Whatman No. 4 paper showed no background, this variety was chosen.

A study was made of the TNB method for 2-ethylhexyl, hydrogen phosphite (BIS), two DB3 methods, and two Schoenemann methods for VX. Briefly, the following were studied:

(1) Variations of zero blank.
(2) Effect of filters on zero blank.
(3) Time stability of color plus effect of heating.
(4) Sensitivity and slope of calibration curves.
(5) Recovery from doped filters.
(6) Evaporation losses from filters.

The information gained from these studies will be useful for the adaptation of chemical procedures for the Auto Analyzer, which will become available in the next quarter.

B. BETA GAUGE TESTS ON AGENT SIMULANTS

The description of a sequential tape sampler modification which automatically determines mass concentration deposited on a paper

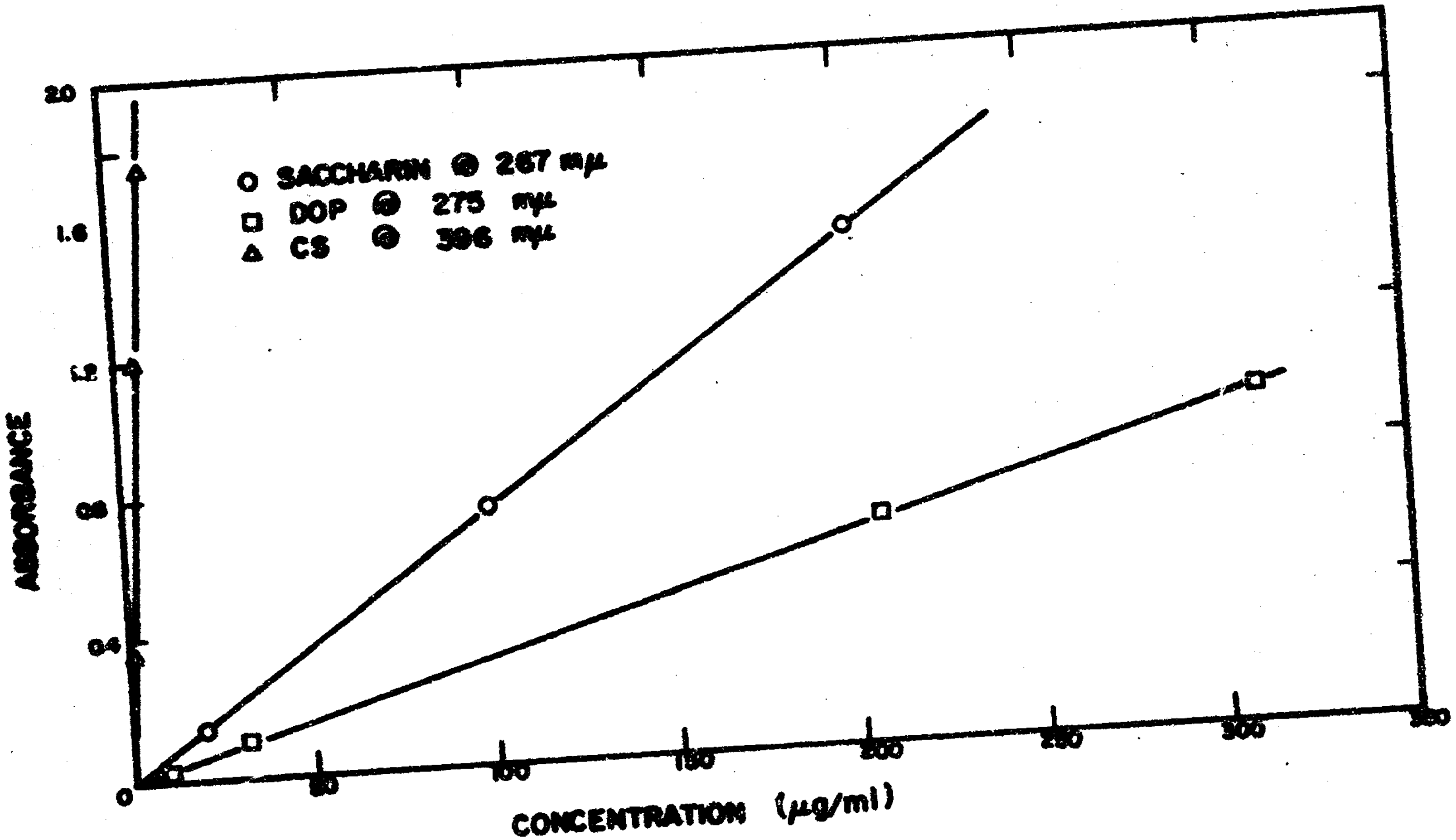

Figure 1. Spectrophotometric calibration curves.

tape has been reviewed in a Harvard School of Public Health report by Bulba and Silverman (Ref. 17). The device consists of a standard AISI tape sampler (as manufactured by RAC Company*), with a modified sampling head containing a one millicurie C^{14} source of β activity and a G-M tube with suitable readout electronics. Because the absorption of β activity depends upon the maximal energy of radiation and the electron density of the absorptive material, β absorption is nearly independent of the type of material present and related only to the amount present. The reason for this is that electron density is proportional to the Z/A ratio (Z = the atomic number, A = the atomic weight). The Z/A ratio for all elements except hydrogen is 0.4 to 0.5. For hydrogen Z/A = 1.

This device has been used for concentrations of the various stack effluents found in open hearth furnace flues. An initial background reading on the substrate (No. 4 Whatman filter paper) is taken and stored in a rudimentary memory bank. This reading is then compared to the attenuated reading after deposition. Although this is done automatically, it can also be done by hand and the comparison carried out manually. The main point is that it does compensate for the variations in substrate material.

This technique appeared to be simple and promising for application to chamber evaluation so that verification was deemed desirable. The following tests were performed with the Harvard equipment on the following aerosols; saccharin, BIS plus carbon black (the carbon black to simulate decomposition products), and pure BIS.

1. <u>Test Procedures</u>

Because it was mainly desired to check the accuracy and sensitivity of the technique (operating ability under automated conditions is well established), cut strips of the substrate were used, rather than the continuous tape. The operating procedure consisted of cutting and weighing the tapes at GCA, transporting them to Harvard where they were tared after being dessicated and an initial β attenuation reading taken. The tapes were then returned to GCA, fitted in a sequential tape sampler and run on varied concentrations of the simulant aerosols. After the tapes were re-weighed to determine the amount of material deposited, they were returned to Harvard for drying, weighing, and final attenuation readings. The results of these tests are presented in Table IV. Figure 2 is a calibration curve for the β gauge. Note that all deposited material weights are transposed to mass per unit area.

---

* Research Appliance Company, Allison Park, Pennsylvania

TABLE IV β GAUGE TEST RESULTS SUMMARY

Test Material	GCA Wgts. w/o humidity cont.				Harv.wgt.w/ humidity cont.				β gauge readings			
	Final wgt. mg	Int. wgt. mg	Δ wgt. mg	Area density mg/cm^2	Final wgt. mg	Int. wgt. mg	Δ wgt. mg	Area density mg/cm^2	final count cpm	Int. count cpm	% Atten.	Area density mg/cm^2
Treated Saccharin	237.6	230.5	7.1	1.4	236.35	228.75	7.60	1.5	9,866	16,650	59.3	1.478
	223.5	220.9	2.6	0.51	222.25	219.25	3.00	0.588	14,138	16,994	83.3	0.519
	224.8	223.0	1.8	0.353	222.60	220.55	2.05	0.403	13,298	15,842	83.9	0.494
	208.8	207.7	1.1	0.216	207.00	205.95	1.05	0.206	16,492	17,694	93.2	0.198
	214.1	213.3	0.8	0.197	212.55	211.55	1.0	0.196	15,520	16,368	95	0.150
Bis	231.2	225.7	5.5	1.08	229.5	224.3	5.2	1.03	12,151	16,882	72	0.928
	227.4	223.6	3.8	0.75	223.95	220.00	3.95	0.788	13,030	17,092	76.7	0.766
	220.9	218.7	2.2	0.434	219.55	217.05	2.5	0.494	16,303	17,692	92.4	0.231
	218.2	217.2	1.0	0.197	216.70	215.40	1.30	0.257	16,958	17,894	94.9	0.166
	218.0	217.2	0.8	0.158	216.35	215.45	0.9	0.198	18,480	18,864	98	0.038
Bis + Carbon	228.8	223.0	5.8	1.14	227.30	221.20	6.1	1.22	13,290	19,558	69	1.091
	225.8	222.3	3.5	0.69	224.45	220.55	3.9	0.77	13,664	16,092	84.7	0.474
	217.4	215.6	1.8	0.355	216.00	213.95	2.05	0.405	17,770	19,218	92.4	0.221
	237.4	236.2	1.2	0.237	236.10	234.30	1.8	0.355	13,192	14,382	90.5	0.283
	216.0	215.4	0.6	0.118	214.35	213.45	0.9	0.177	16,724	18,210	91.8	0.241
Bis + Carbon + Carbon Dusted on Filter	224.7	223.3	1.4	0.276	223.15	221.55	1.6	0.316	16,672	17,794	93.5	0.194

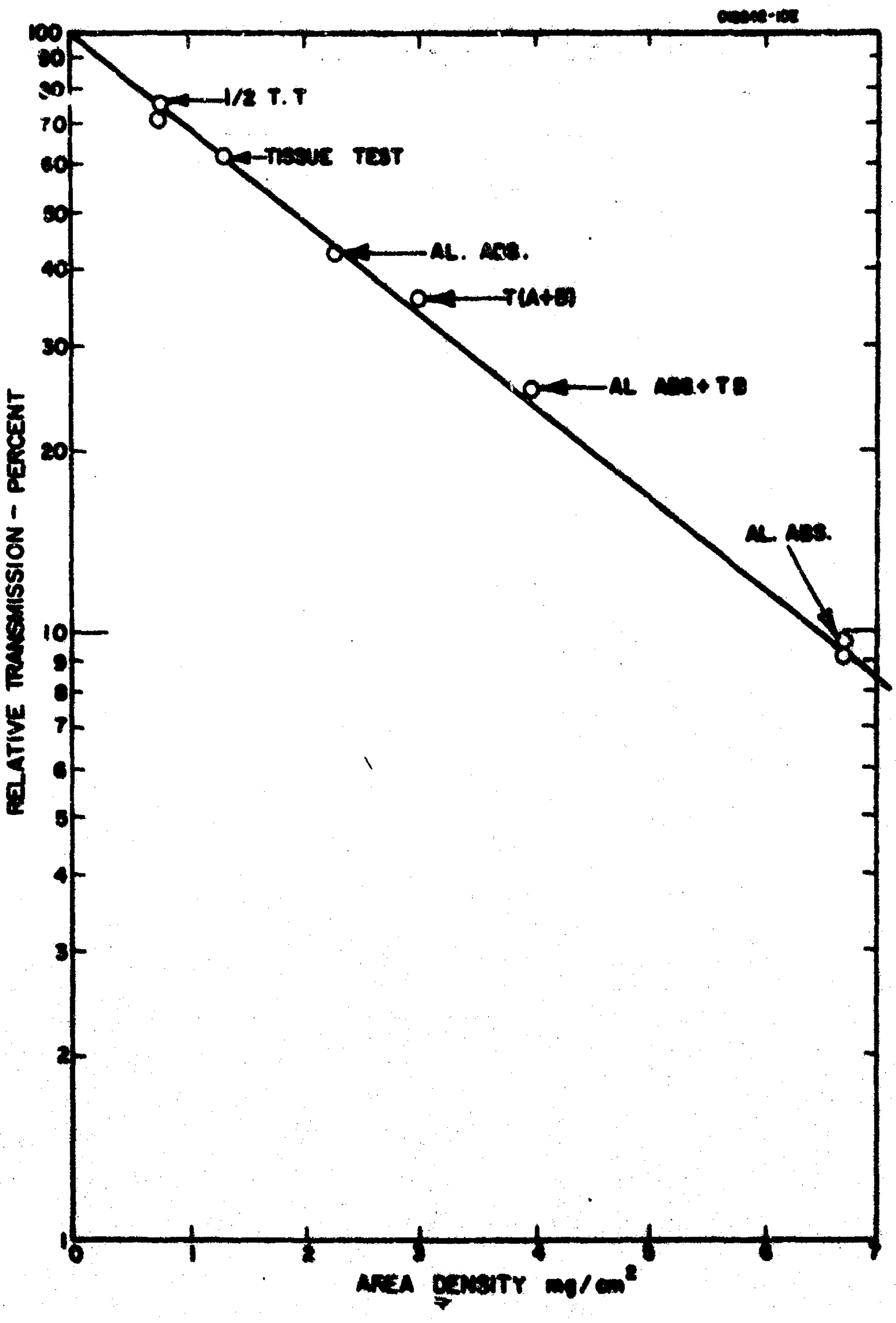

Figure 2. Calibration curve.

The details of the test set up and calculations for the equipment at Harvard are given below, approximately as received.

Beta Gauge Calibration

Source: 1 mCi C^{14} prepared in aluminum disc 1" diameter

Target: Blank filters Whatman No.41 average density 9.45 mg/cm^2, and samples collected on pre-weighed, pre-counted blanks.

Counter: Amperex G-M tube, 1.4 mg/cm^2 mica window, 1,400 V operating voltage

Scaler: Baird Atomic Multiscaler II M 132.

Counting Statistics:

Taken on Blank No. 1.

Count No.

1.	17,138 cpm	6.	17,282
2.	17,328	7.	17,520
3.	17,186	8.	17,210
4.	17,350	9.	17,350
5.	17,462	10.	17,465

$$\sigma = \sqrt{n} = 132 \text{ cpm}$$

Expected deviation at 95 percent probability $\pm 2\sigma = \pm 264$

Calibration curve: (See graph, Figure 2)

Absorber	Mass per unit area
1/2 Paper tissue A	0.675 mg/cm^2
Paper tissue A	1.35
Paper tissue B	1.65
Aluminum A	2.30
Paper tissue A + B	3.00
Paper tissue B + Alum. A	3.95
Aluminum B	6.7

2. Discussion

The purpose of weighing at both GCA and Harvard was for control and to determine if dessication had any effect or was at all necessary. From an inspection of Table IV, it appears that this procedure was not entirely necessary and would not be a factor in the operation of the unit, although during calibration it is worthwhile. The β gauge readings, when interpreted with the calibration curve furnished indicate excellent results for treated saccharin and also for BIS with somewhat poorer agreement on BIS plus carbon. The amount of carbon added to the BIS was not precisely monitored and there is a problem with the liquids spreading out and saturating the paper beyond the area inspected by the β gauge. This spreading occurs rather slowly and would not be a problem in an automatic instrument. The greatest accuracy with this technique is achievable only when the β transmission is below a value of approximately 70 percent. For the system evaluated here, this corresponds to 1 mg/cm^2 or approximately 5 milligrams for a 1-inch diameter spot, which would be a most convenient size to use if a normal tape sampler were to be converted. Assuming a maximum desired sampling time of 1 minute and a maximum attainable flow rate of 30 ℓ/min, the minimum concentration which would be detectable with this apparatus is 167 micrograms per liter (approximately 0.2 gm per M^3). The result is discouraging and it constitutes poor sensitivity for chamber testing since our experience indicates a minimum required sensitivity of 5 μg/liter.

The area densities (mg/cm^2) found with the Harvard β gauge are compared with values calculated by weighing the filters on a Mettler analytical balance before and after testing (Figure 3).

C. POROUS ELECTRODE PRECIPITATOR

The Porous Electrode Precipitator (Ref. 14) is designated Unit MB and was built for the Space General Corporation by Metronics Associates, Inc., Palo Alto, California. The basic flow for the unit is 1000 l/min. The particulate matter contained in this air stream is electrostatically precipitated onto a porous electrode through which a stream of treated water flows at 5 cc/min, i.e., the contents of 1,000 liters of air per minute are concentrated into 5 cc of water per minute.

The collection efficiency for 1.3 micron polystyrene latex beads is reported as being better than 88 percent when determined in the usual manner. The water was treated with 4 grams of NaCl so that it would be conductive and 0.5 cc of Photo Flow was added to each filter so that surface tension would be lower for flow through the porous frit.

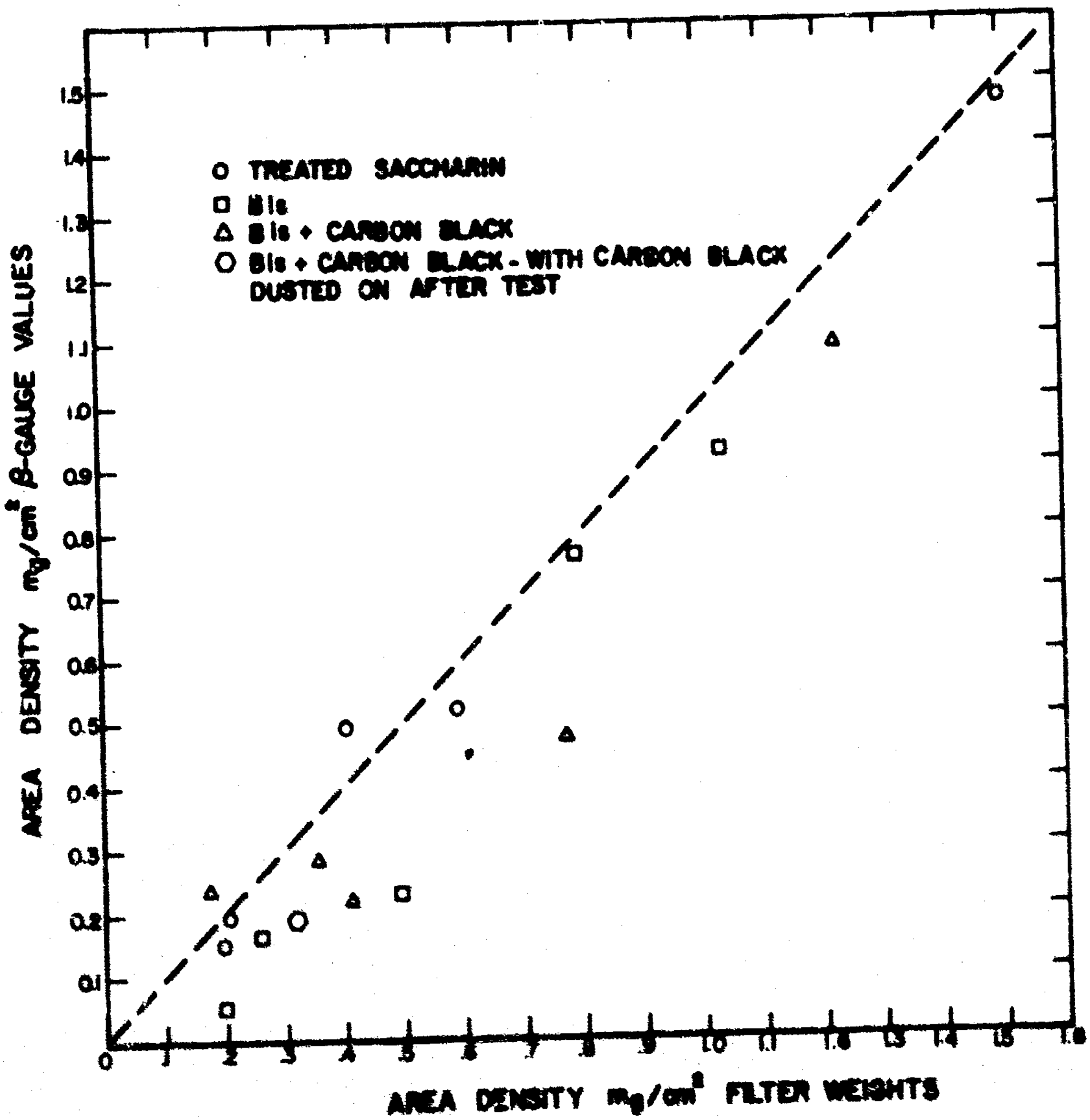

Figure 3. Correlation between β gauge and gravimetric measurements.

Since the concentration ratio of this device is favorable, a letter was written to Metronics to determine its availability. A reply dated 15 Fabruary 1966 and signed by G. Harman, Marketing Manager, indicated that such a device was indeed under study by them, but it was not available for sale.

Since an immediate assessment of the promise of this device was needed, a crude cylindrical approximation of it was constructed from the report description (Figure 4). Based on preliminary tests with this precipitator and from a perusal of the Metronic's report, it can be said that the device appears to merit inclusion in our test program with the appropriate geometric and electric optimization. Therefore, it is recommended that an appropriate device be designed and constructed for initial evaluation in the cubic meter test chamber and that it be sufficiently scaled down to be compatible with the liquid flow rates required for the Technicon device.

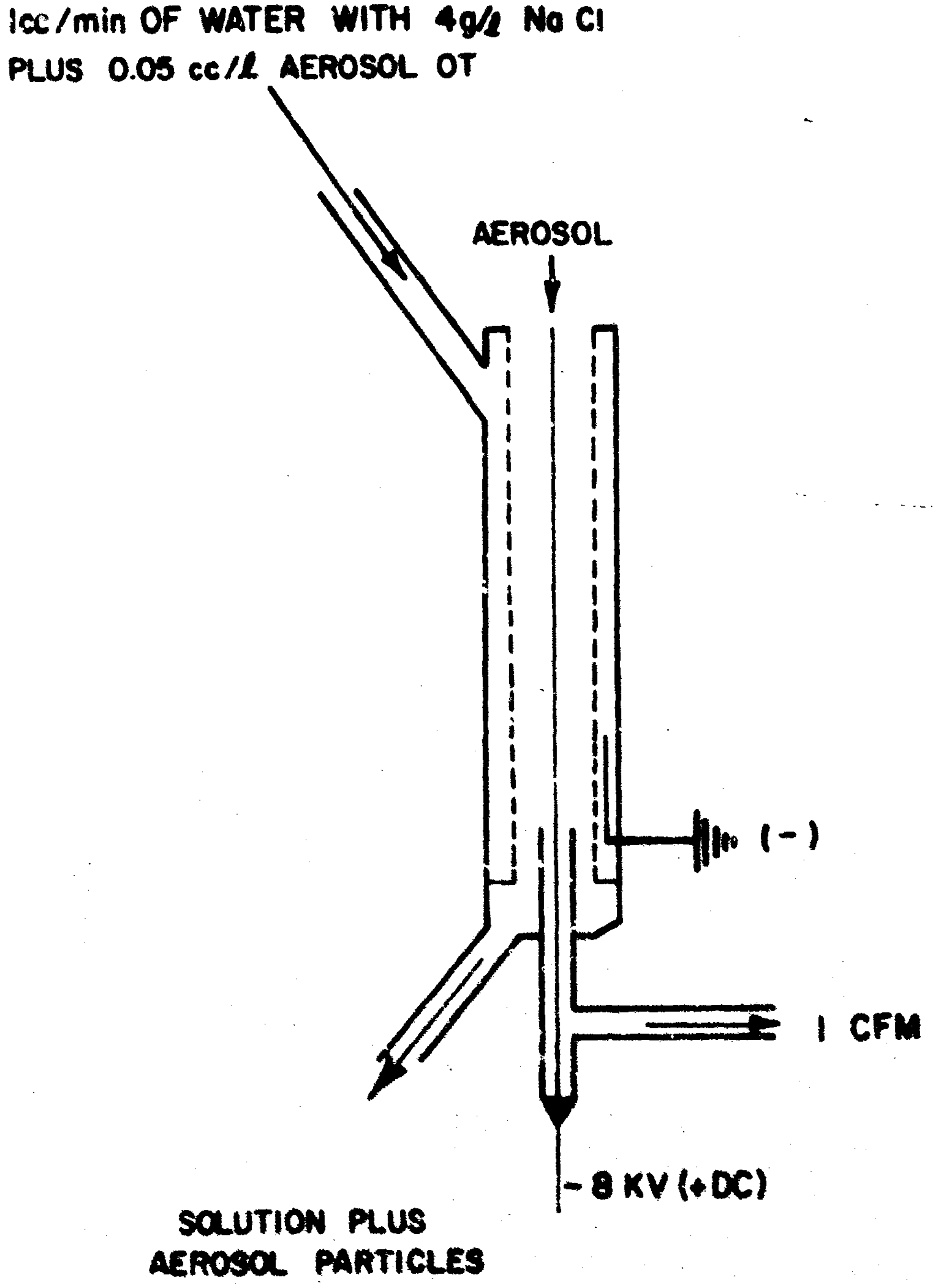

Figure 4. Porous electrode precipitator.

D. TEST RESULTS

1. Chamber Floor Sampling

Several tests were run using fairly monodisperse 12μ special co-polymer plastic spheres as a test aerosol. There is no known chemical analysis for this material and it is rather costly ($10/gm) to use in quantities large enough for weight analysis. The sampling technique decided upon was a combination of membrane filter samples, cascade impactor, Royco, and coated floor slides.

Filter samples were taken at 0.5 min intervals for 0.1 min on the membrane filters for 6 min after dissemination. A cascade impactor sample was taken at 2.0 min intervals for 0.1 min. The stages of the impactor were coated with 50 centistoke silicon oil. Royco counts were taken for 15 min after dissemination. The Royco sample was taken from scans of 8, 10, 12, and > 12μ channels alternately. Ten, 1- by 3-in. glass microscope slides coated with 50 centistoke silicone oil were placed symmetrically on the chamber floor (Figure 5) in order to collect fallout.

The membrane filter samples were counted using a light field microscope. The procedure employed was to count representative areas of each filter, noting the number of single particles and the number of groups of particles and then noting whether the grouped particles formed chains or clusters. The total particles counted was prorated to give the number of particles present on the entire filter. From a volume diameter cubed (D_v^3) of 2852 (which was found by sizing the spheres prior to testing) and a density of 1.2 g/cc (which was supplied by the manufacturer) the total weight of material on each filter was determined. Mass depletion curves (Figure 6) were plotted for both tests. The tests were dissimilar in that one showed a very rapid depletion (792 mg/m^3 at 0.5 min to 32 mg/m^3 at 6 min) while the other test indicated a decrease in the rate of decay of concentration after 4 min. In this test, the concentration fell from 548 mg/m^3 at 0.5 min to 126 mg/m^3 at 6 min. The area counted on each filter varied according to the particle density on the filter. Counted areas ranged from as little as 2 percent of the area in the first filters up to 6.5 percent of the area in some of the sparser filters. Statistically, at least 10 percent of the filter area should be scanned in order to obtain any high degree of reliability of the data.

Several photo-micrographs were taken of each membrane filter under a 100X magnification and an analysis of the agglomerate distribution was made, (Table V).

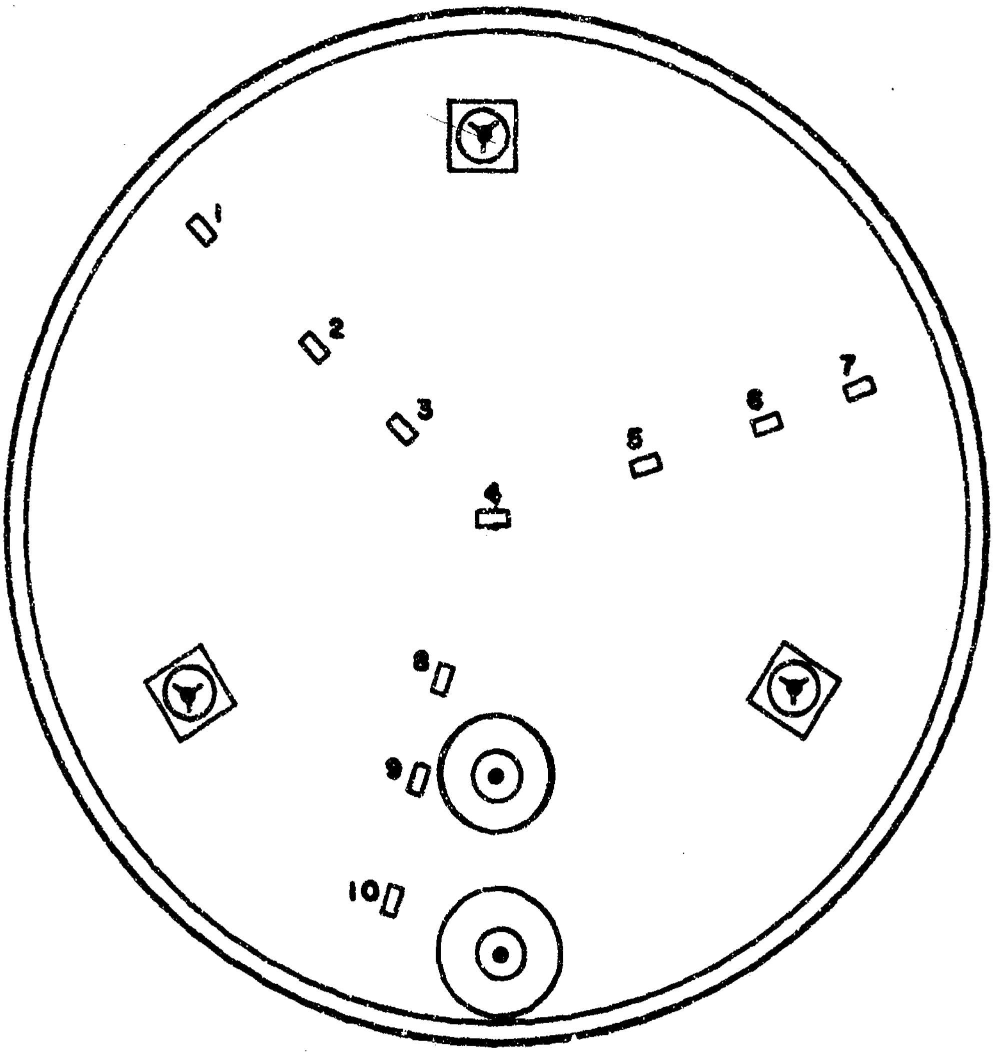

Figure 5. Position of slide fallout collectors.

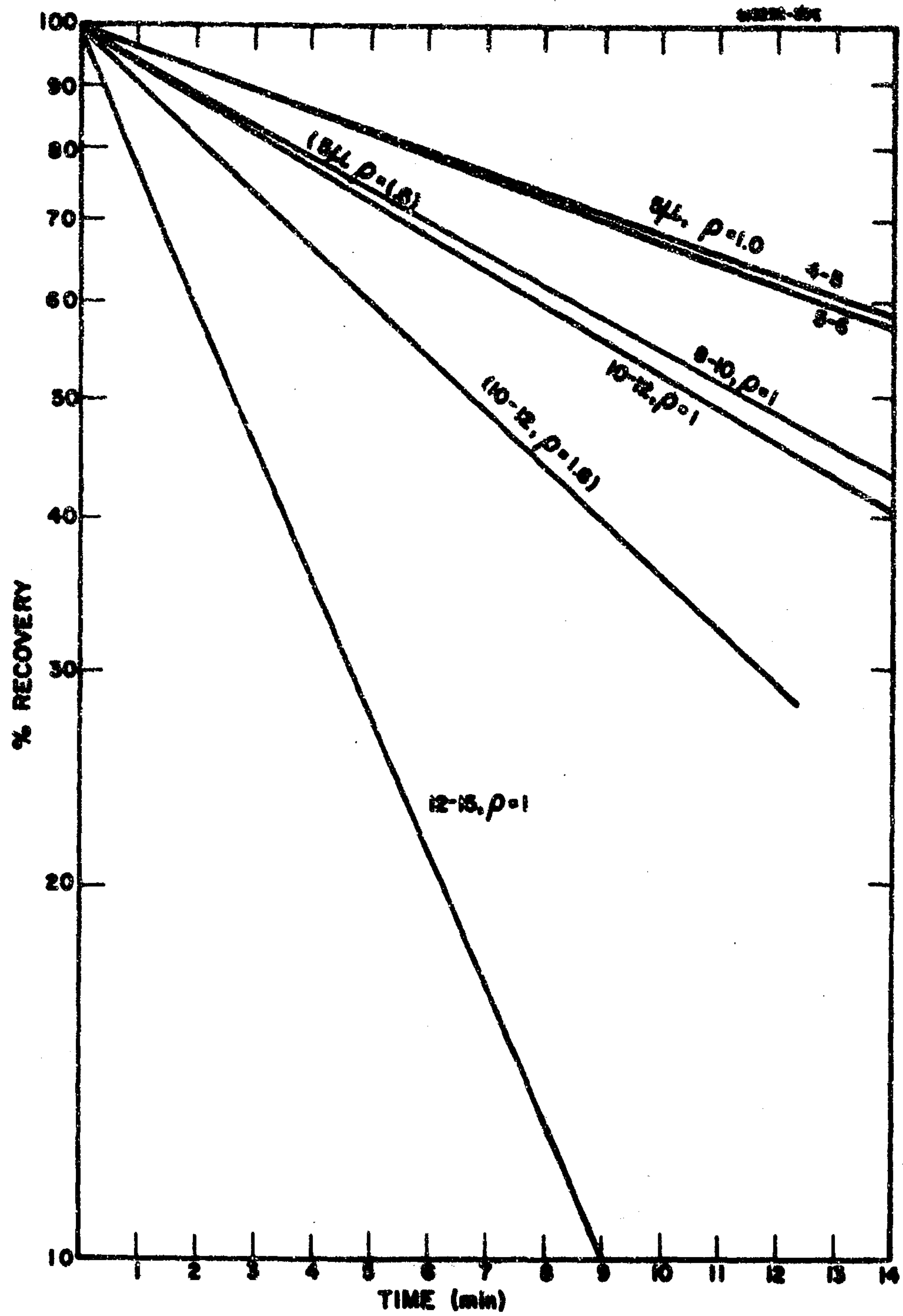

Figure 6. Mass depletion curves.

TABLE V

Particles in Agglomerate	Number Frequency	% by Count	Total Particles
1	113	78.5	113
2	21	14.6	42
3	8	5.5	24
4	1	0.7	4
7	1	0.7	7
	144	100.0	190

The analysis of the data shows that 78.5 percent of the particles (considering an agglomerate as a single particle) are present as singlets. If the distribution is treated as a monodisperse system, 59.5 percent of the total mass appears as discrete particles.

The floor slides were counted using the same technique employed for counting the membrane filter samples. However, there were many multi-particle agglomerates on the slides. The agglomerates were grouped by the number of agglomerates and the number of particles in each agglomerate. The total of all particles counted on each slide was prorated from the area counted to the total number of particles on the slide. The prorated average on each slide was approximately 10^6 particles. The total particles on all the slides was 8.9×10^6 and 8.7×10^6 for the two tests run. All particles on all the slides were prorated to find the number of particles on the chamber floor. Using this value and the values for D_V^3 and density previously employed, the total mass of particles on the floor was found to be 1.34g and 1.30g for the two tests run. One gram of aerosol was disseminated for both tests. The variation in the weight calculated to be on the chamber floor and the weight expected to be present probably is due to the fact that less than 1 percent of the slide area was counted and all the slides together only comprise 2 percent of the floor area. It is clear that the use of floor slides to determine accountability of the aerosol used is rather an involved and inaccurate method unless a sufficient area of the floor is covered and a more convenient method for analysis of the slides (chemical) is employed. The only other method of analysis available for this aerosol is by weight and this was not practical for the amounts of material collected.

The cascade impactor samples for these tests were disappointing, as is often the case when sampling a monodisperse aerosol with

the cascade impactor. Only one of the tests produced an impactor sample which was countable. The mass median diameters on the individual stages were 16.3, 19.5, and 13.1μ for the first three stages. There were no particles on the fourth and fifth stages. The first and second stages appeared to have no sharp cutoff between the point of heaviest particle concentration and the edge of the glass plate. The third stage did have a defined cutoff between the place of maximum particle concentration and the edge of the plate, but an oblong portion of the plate in the center was void of particles. The procedure used to size the particles was to chose an area and count and size the particles in the area considering any agglomerates in toto. The discrepancy between the mass median diameters found on the first and second stages may be explained by the fact that there were more agglomerates noted on the second stage than on the first stage since the particles in the agglomerates were not counted as individual small particles, but as aggregate large particles. (See Table VI) It is reasonable to assume a resulting larger mass median diameter for the second stage.

TABLE VI

Stage	Singles	Doubles	Particle Groupings Triples	Fours	Fives	Sixes
1	264	33	5	1		
2	175	50	30	9	4	1

Royco counts were taken for both tests, but the counts were so low that no further analysis of the data was made except to indicate that the maximum particle counts occurred in the 10 to 12μ channel. This is consistent with the manufacturers indicated count median diameter of 12μ. The membrane filter downstream of the Royco viewing volume was sized to check the manufacturers claims concerning size. The results of a 300 particle count indicate a count median diameter (Mg) of 13.2μ, a mass median diameter (Mg′) of 14.6μ and a standard geometric deviation (σ_g) of 1.1.

2. Automatic Particle Counter Material Balance

Early in the testing period a preliminary test was performed to compare the Royco counter with particles decaying from the atmosphere onto drop slides. One gram of treated saccharin was dispersed into the cubic meter chamber using the dry powder ejector. After a suitable counting period the slides were removed simultaneously with the last counted channel on the Royco.

The drop slides were sized with a light field microscope and the data from the slides as well as the counter was processed through the computer. The mass median diameters extracted from the slides were 9.5μ and 12.0μ while the Royco mass median diameters were 11.7μ and 12.0μ respectively, which shows good agreement, considering the built-in error due to length of time needed for the Royco to scan a complete size range.

3. Particle Decay Analysis

One of the advantages of having a small test chamber is the short periods one has to monitor particle decay. For example, one gram of treated saccharin was generated into the chamber using the dry powder ejector and one particular size range monitored on the automatic particle counter. Each channel was sampled for 0.32 min and the counts recorded. The next count was of all particles greater than the size range of interest to enable a coincidence correction to be made. Five individual size ranges were monitored in five separate tests. The results are given graphically in Figure 7. Further utilization of the Royco will be made in this area of study.

4. Removal of Interference Particulates From Latex Sphere Aerosols

To calibrate the Royco PC 200 automatic particle counter, it is necessary to introduce monodisperse spheres of known size directly into the instrument. A homogeneous polystyrene latex sphere generator constructed for this purpose performs this function adequately. However, along with the required size spheres a high background of particles are recorded in the 0.3μ channel. Since this high background should not enter the counter, it has been proposed to charge the exiting particles from the generator with an α emitter (Polonium Source) and thereby precipitate the background particulates while letting the larger particles with higher mass and momentum pass through into the counting cell. Preliminary testing of this method is encouraging and it will be investigated further.

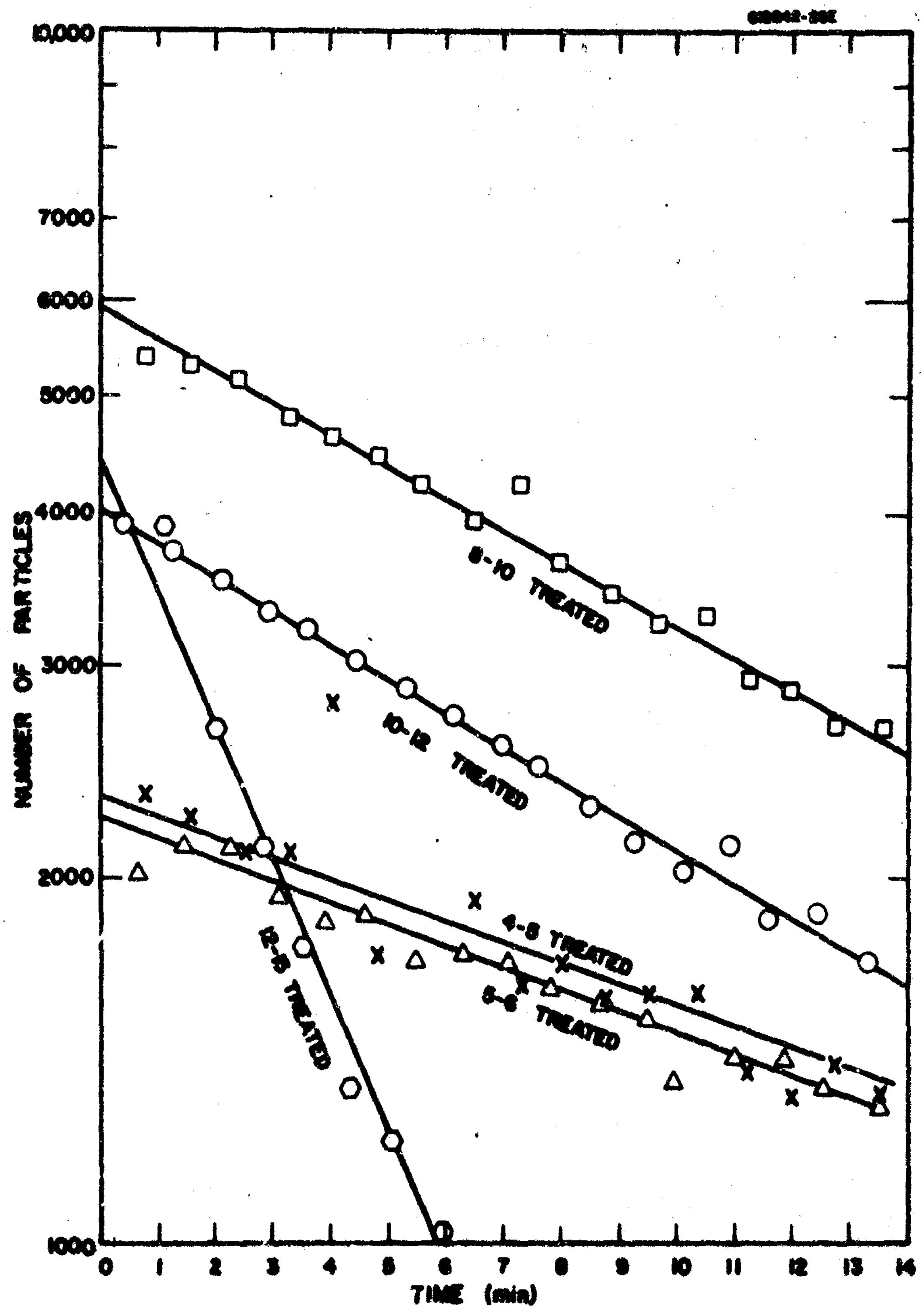

Figure 7. Individual size ranges of treated samples.

5. Viewing Microscope

While analyzing the tests which used monodisperse spheres as the aerosol, it was apparent that agglomeration of the particles posed a serious problem. The percent agglomeration before reaching the floor and the percent of agglomeration on the floor due to one particulate falling onto or next to another was not known. To resolve this problem, an optical microscope was fitted with a 2-in diameter, 4-in long tube and bracketed in an inverted position (Figure 8). A glass slide was placed on the end of the tube which was inserted into the bottom of the test chamber. This enables the manner in which the particle fallout occurs to be observed, i.e., single particles depositing as single particles, single particles depositing and agglomerating, agglomerates depositing as agglomerates and agglomerates breaking up on impaction into smaller agglomerates or singles. A Siemes camera records the manner in which the particles deposit.

6. Mass Decay of Saccharin Aerosols

Use of mass decay curves frequently enables investigators to determine the approximate mass median diameter of the decaying aerosol by the best fit method. These theoretical curves are plotted for a spherical aerosol whose density is 1.0 gm/cm^3.

In using saccharin as a test aerosol with this method, it was impossible to evaluate the aerosol cloud because: (1) the shape factor and (2) the apparent density were both underdetermined.

Several tests were performed taking simultaneous cascade impactor and membrane filter samples to determine the true particle diameter. These two values compared rather nicely, but when the transparent curve analysis was applied to the decay curves a large deviation was observed. This large deviation indicated that it would be necessary to determine the saccharin shape factor and apparent or effective particle density.

Masswise, three methods of analysis compared very well (Table VII) (1)1106B all glass filter samples, (2) cascade impactor total mass collection, and (3) automatic particle counter analysis. It is realized that there is an inherent error in the automatic particle analysis at any individual time, but since this analysis as well as the cascade impactor analysis were both made over the same time range, the deviation due to time decay from the actual values was minimized. The comparison between these values and the filter samples (which were also made over a similar time range) is considered significant because of the: (1) indication of little or no interstage deposition in the cascade impactor, (2) relatively even particle distribution at several points in the chamber indicating thorough mixing, and (3) reliability in filter sampling from a stirred chamber.

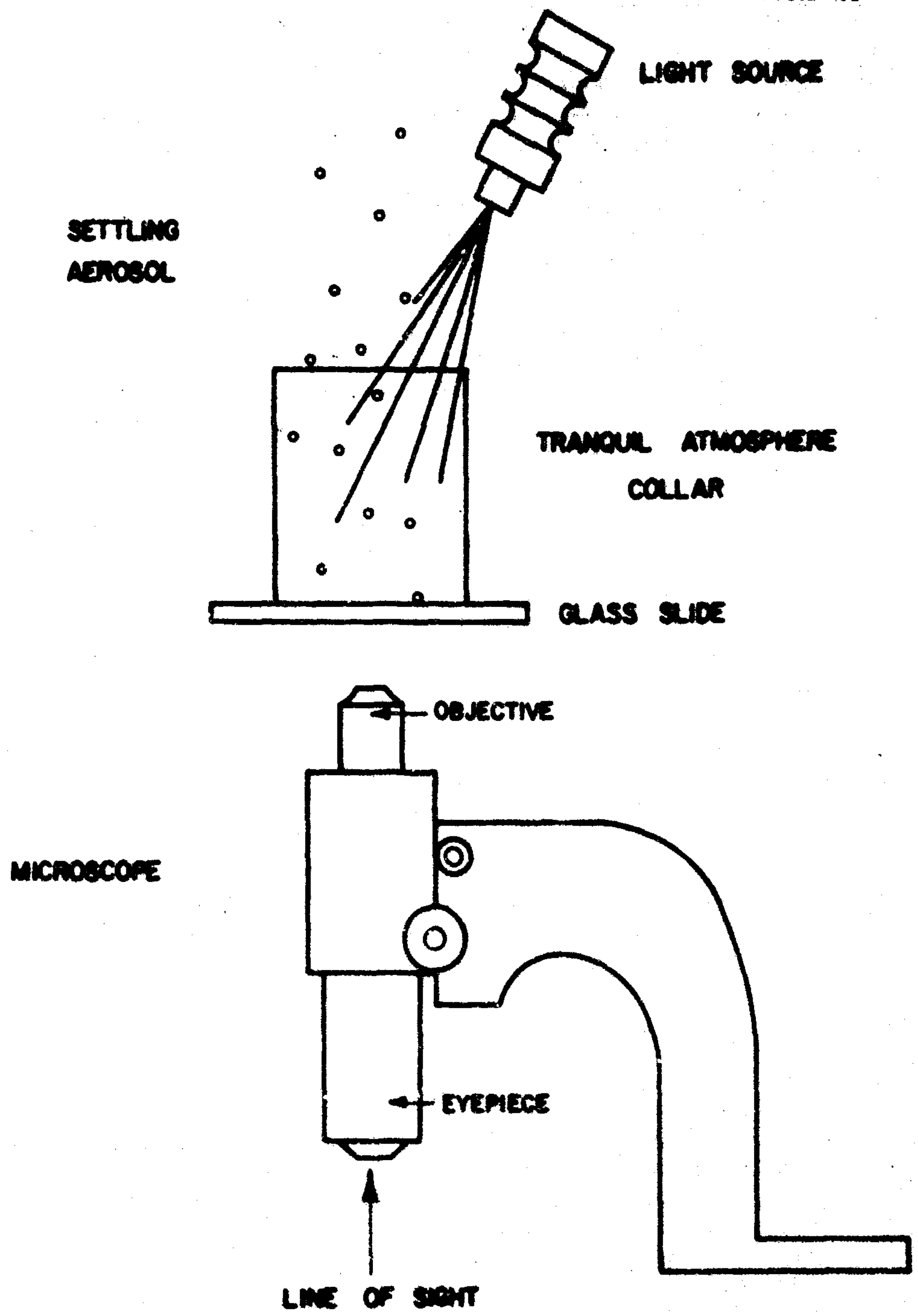

Figure 8. Viewing microscope.

V. PROGRAM FOR THE FIFTH QUARTER

During the fifth quarter it is planned to do extensive study on chamber mixing. It is very important for aerosol assessment work that the chamber have a uniform concentration.

The variables which will be studied are: (1) number of fans, (2) fan height, (3) fan orientation, and (4) fan speed.

It is realized that even though a particular configuration may be found which will give the desired mixing characteristics, it may not apply to different chambers; nevertheless, it is a much needed experiment if a proper evaluation of the aerosol cloud and the instrumentation is to be accomplished.

The GCA "in-line" cascade impactor will be calibrated using DOP as an aerosol. Much literature is available on the use of this substance as an aerosol and it has the advantage of having a spherical shape when generated and a density near unity (0.99 gm/cm^3) and can therefore be evaluated by the use of the mass decay transparency curves. New dissemination methods will be tested in an attempt to generate a DOP aerosol of proper size (M_g' - 15 microns) and of proper concentration (~ 500 mg/cm^3 to simulate conditions in the large test facility) in a short time period (0.05 minutes).

As part of the continuing effort considered essential to the testing program for the chamber are: (1) determination of optimum geometry, orientation, and sampling velocity for each sampling nozzle; and (2) investigation of aerosol charge properties and on cloud assessment. These investigations will continue.

REFERENCES

1. Dennis, et al., R., "Aerosol Dissemination Assessment". First Quarterly Report for U.S. Army Edgewood Arsenal, Contract DA-18-035-AMC-376(A), GCA Technical Report No. 65-22-G, (December 1965).

2. Dennis, R. and Hommel, C.O., "Aerosol Dissemination Assessment". Second Quarterly Report for U.S. Army Edgewood Arsenal, Contract DA-18-035-AMC-376(A), GCA Technical Report No. 66-5-G (May 1966).

3. Dennis, R. and Gordon, D., "Aerosol Dissemination Assessment". U.S. Army Edgewood Arsenal, Contract DA-18-035-AMC-376(A), GCA Technical Report No. 66-20-G (October 1966).

4. Dimmick, R.L., Hatch, M.T., and James, N.G., "A Particle-Sizing Method for Aerosols and Time Particles". A.M.A. Arch. of Ind. Health, 18, 23 (July 1958).

5. Gilbert, T.L. and McCully, C.R., "The Determination of Polydisperse Aerosol Size Distribution from the Analysis of Light Scattering Data". Armour Research Foundation, Scientific Report No. 12 (20 August 1954).

6. Talley, R.L., "Recovery of Approximate Aerosol Distributions by Direct Inversion of Settling Curves". Chord Memo 603, Cornell Aeronautical Laboratory, Ind. (1965).

7. Twomey, S. and Severynse, G.T., "Measurements of Size Distributions of Natural Aerosols". J. Atmos. Sci. 20, 392 (September 1963).

8. Twomey, S., "On the Numerical Solution of Fredholm Integral Equations of First Kind of Inversion of the Linear System Produced by Quadrature, J. Assoc. Comp. Mach., 10, 97 to 101 (1963.

9. Twomey, S., "The Determination of Aerosol Size Distributions from Diffusional Decay Measurements". J. Franklin Inst., 275, 121 to 138 (1963).

10. Shifren, K. and Perelman, A., "Calculation of Particle Size Distribution by Data on Spectral Transparency". Pure and Applied Geophysics, 58, II, 208 to 220 (1964).

11. Phillips, D.L., "A Technique for Numerical Solution of Certain Integral Equations of the First Kind". J. Assoc. Comp. Mach., 9, 84 to 97 (1962).

REFERENCES (continued)

12. "New Techniques for Dissemination of Chemical Agents", Annual Summary Report, Physical Research Laboratory, Research Laboratory, Edgewood Arsenal, Maryland. Contract DA-18-108-AMC-249(A), GCA Technical Report Number 66-14-G (September 1966).

13. Lundgren, D. and Graf, L., "Development of an Aerosol Collector for Use in a Microbiological Aerosol Detection System". Contract DA-18-064-CML-284(A), Litton Systems, Inc. (October 1964).

14. Space General Corporation, "Research Program on BW Detection". Fifth Comprehensive Report, AGC 382-7, Vol. 1, p. 3 to 41, Contract DA-18-064-AMC-137(A). (April 1965).

15. May, K.R., "The Cascade Impactor". J. Sci. Instr. (London) 22, 187 (1945).

16. Levin, L.M., "The Intake of Aerosol Samples". Translation by K. Syers fu Izv. Akad Nauk, SSSR Ser. Geofis No. 7, 914 to 925 (1957). Reprint Order No. CEO 112.

17. Bulba, E. and Silverman, L., "A Mass Recording Stack Monitoring System for Particulates", presented at the 58th APCA Meeting at Toronto, Canada. (June 20 to 24, 1965).

DISTRIBUTION LIST

11 copies to	US Army Edgewood Arsenal Research Laboratories ATTN: Donald E. Buck, Edgewood Arsenal, Maryland 21010
20 copies to	Defense Documentation Center Headquarters ATTN: DDC-IRS, Cameron Station, Alexandria Virginia 22314

DOCUMENT CONTROL DATA - R&D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY *(Corporate author)* GCA CORPORATION GCA TECHNOLOGY DIVISION Bedford, Massachusetts	2a. REPORT SECURITY CLASSIFICATION UNCLASSIFIED 2b. GROUP N/A

3. REPORT TITLE

AEROSOL DISSEMINATION ASSESSMENT

4. DESCRIPTIVE NOTES *(Type of report and inclusive dates)*

First Annual Summary Report June 1965-June 1966

5. AUTHOR(S) *(Last name, first name, initial)*

Dennis, R Gordon, D. Hommel, C.O.
Doyle, A.W. Gussman, R.A. Sacco, A.M.

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
April 1967	49	17

8a. CONTRACT OR GRANT NO. DA-18-035-AMC-376(A) b. PROJECT NO. 1B522301A081 c. TASK d.	9a. ORIGINATOR'S REPORT NUMBER(S) GCA-TR-67-2-G 9b. OTHER REPORT NO(S) *(Any other numbers that may be assigned this report)* N/A

10. AVAILABILITY/LIMITATION NOTICES This document is subject to special export controls and transmittal to foreign nationals or foreign governments may be made only with prior approval of the CO, Edgewood Arsenal, ATTN: SMUEA-TSTI-T, Edgewood, Maryland 21010.

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY
Chemical agent dissemination	Physical Research Laboratory RESEARCH LABORATORIES

13. ABSTRACT The basic program objectives are to develop, to design, and to fabricate a complete aerosol assessment system. These were reviewed in terms of the capability of the instrumentation for the measurement of the mass concentration of active agent, concentration of degraded agent and inert materials, describing particle-size parameters for active agents, shape factor for solid particles, and bioeffectiveness. Sampling techniques were reviewed; e.g., filtration, inertial collection, scrubbing and absorbing, electrostatic, thermal percipitator, and sedimentation, interpreted in terms of their applicability to the system. Special attention was given to proposed instruments for particle-size assessment. Several analytical methods for mass assessment are discussed including wet-chemical techniques and instrumental methods. Results of experiments in a 1 m^3 test chamber are presented and discussed in such terms as floor sampling, an automatic particle counter, a beta absorption mass meter, a porous electrode collector, particle decay analysis, a viewing microscope, mean decay of saccharin aerosols, sample size, location and time, and representative sampling.

14. KEYWORDS: Aerosol assessment
Particle size
Aerosol sampling
Aerosol-collection techniques
Analytical methods
Absorption

DD FORM 1 JAN 64 1473